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Ground state hyperfine structures of ^{43}K and ^{44}K measured by atomic beam magnetic resonance coupled with laser optical pumping

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Résumé. — Les structures hyperfines de l'état fondamental de ^{43}K et ^{44}K ont été mesurées par une méthode de résonance magnétique sur jet atomique dans laquelle les spins atomiques sont orientés par pompage optique laser. Les résultats spectroscopiques sont les suivants :

$$\Delta\nu_{43}(^2S_{1/2}) = 192{,}648\,4\,(30)\ \text{MHz} \quad \text{et} \quad \Delta\nu_{44}(^2S_{1/2}) = -\,946{,}718\,(3)\ \text{MHz}\,.$$

La sensibilité de notre méthode est comparée à celle obtenue avec un appareil correspondant au dispositif classique de Rabi.

Abstract. — The ground state hyperfine structures of ^{43}K and ^{44}K have been measured by an atomic beam magnetic resonance method in which the atoms are spin-polarized by laser optical pumping. The spectroscopic results are : $\Delta\nu_{43}(^2S_{1/2}) = 192.648\,4\ (30)$ MHz and $\Delta\nu_{44}(^2S_{1/2}) = -\,946.718\ (3)$ MHz. The sensitivity of our method is compared to the one achieved in classical ABMR apparatus.

1. **Introduction.** — High resolution laser spectroscopy on long series of isotopes has shown to be a very fruitful method for the investigation of nuclei properties [1-3]. A wealth of data has been obtained in this way for Na [4], Rb [5], Cs [6], Fr [7] and very recently for K isotopes [8]. Isotope shifts (*I.S.*), nuclear spins (I), hyperfine constants of the ground state [$A(^2S_{1/2})$] as well as of the excited states [$A(^2P_{1/2})$, $A(^2P_{3/2})$, $B(^2P_{3/2})$] have been determined by optical spectroscopy, with an accuracy of a few MHz, even 1 MHz in the best cases.

In a recent experiment [9] on the D_1 and D_2 lines of ^{39,40,41}K using well collimated atomic beams and modulated laser light, an accuracy and a reproducibility of a few hundred kHz for the frequency intervals measurements have been obtained. This is comparable to the usual precision given by radio-frequency methods for the same excited states (optical double resonance and level crossing) [10-12].

On the other hand, for the hyperfine structure of the ground state, it is well known that the r.f. spectroscopy [13] leads to much more accurate measurements than the optical spectroscopy : the levels involved in the r.f. transitions are hyperfine sublevels of the ground state ; the frequency width of the resonances is then only limited by the interaction time. With sophisticated techniques, precisions better than 1 Hz have been reported [14, 15] and it is possible, without special care, to get an accuracy of a few kHz which is already three orders of magnitude better than that obtained in optical laser spectroscopy.

In the present paper we report on an r.f. magnetic resonance experiment on radioactive potassium isotopes performed during the laser optical spectroscopy experiment quoted above and using the same apparatus. As a test of the accuracy obtained with this experimental set-up, the well known h.f.s. of ^{45}K and ^{20}Na have been measured. Then the h.f.s. of ^{43}K and ^{44}K have been determined. The h.f.s. of other

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isotopes have not been studied in this way because of the frequency limitation of the r.f. amplifier (1 GHz).

2. **Principle of the experiment.** — The principle of the laser spectroscopy experiment has been extensively explained in previous papers [4, 5, 16]. In brief, a thermal atomic beam of potassium interacts at right angle with the light beam from a C.W. tunable single mode dye laser (see Fig. 1*a*) in the presence of a weak static magnetic field H_0, which defines the quantization axis, parallel to the laser beam. At optical resonance with one of the hyperfine components of the D_1 line

$$(4\,^2S_{1/2} \leftrightarrow 4\,^2P_{1/2}),$$

the laser light induces an optical pumping which changes the population distribution between the magnetic substates (F, m_F) of the ground state. These population changes are analysed by a sixpole magnet, which focusses the atoms corresponding to $m_J = +1/2$ and defocusses those corresponding to $m_J = -1/2$ [16]. After the sixpole magnet, the focussed atoms of the beam are ionized, mass separated and counted. When the laser frequency is scanned, the optical resonances appear as positive or negative peaks on a baseline ion signal. One can obtain in this way a first determination of the hyperfine ground state splitting with an accuracy of a few MHz. Moreover, the sign of the nuclear magnetic moment can be deduced from the relative intensities of the four components of the hyperfine structure.

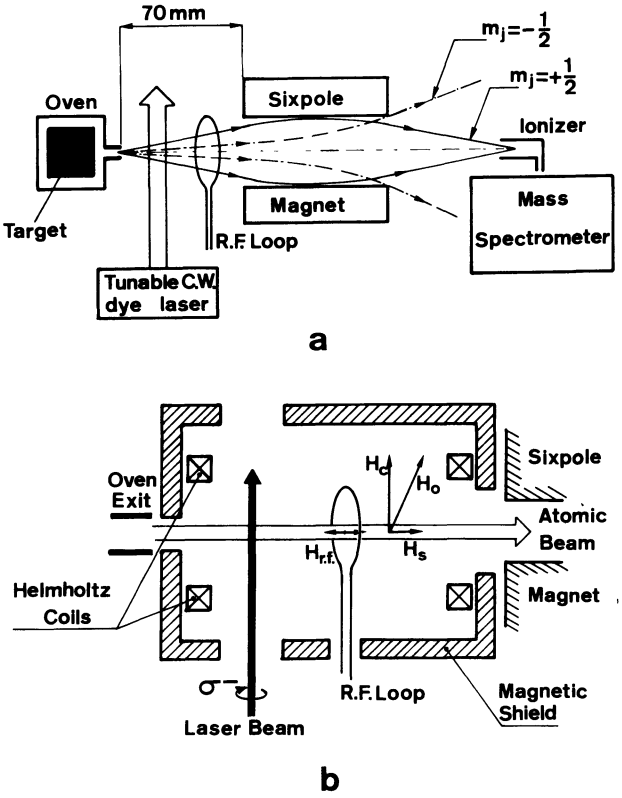

Fig. 1. — *a*) Principle of the experiment; *b*) Schematic view of the interaction region.

Let us now suppose that the laser frequency is locked on the frequency of the atomic transition giving the strongest negative peak, i.e. for an isotope with positive nuclear magnetic moment

$$4\,^2S_{1/2}\,F = I + 1/2 \leftrightarrow 4\,^2P_{1/2}\,F' = I + 1/2$$

or in the case of negative nuclear magnetic moment

$$4\,^2S_{1/2}F = I - 1/2 \leftrightarrow 4\,^2P_{1/2}\,F' = I + 1/2\,.$$

The laser light beam is σ^- polarized with respect to H_0. Under these conditions, the detected atomic beam intensity is strongly reduced since, by optical pumping, the atoms are transferred from the focussed $F = I + 1/2$ (or $F = I - 1/2$ for $\mu_I < 0$) state to the defocussed state $F = I - 1/2$ (or $F = I + 1/2$ for $\mu_I < 0$). Between the interaction region with the laser light and the sixpole magnet, a r.f. magnetic field given by a r.f. loop is applied to the optically pumped atoms (Fig. 1*a*). When the r.f. frequency is set equal to the frequency of an allowed magnetic transition between two Zeeman sublevels of the two ground state hyperfine levels, the r.f. magnetic field tends to equalize the populations of the involved sublevels, so that the observed signal increases again. Since H_0 is known, the hyperfine interval in zero field can be deduced from the frequencies of the observed magnetic transitions $F = I - 1/2, m_F \leftrightarrow F = I + 1/2, m_F + \Delta m_F$ where :

$\Delta m_F = \pm 1$ if the r.f. field is perpendicular to H_0.

$\Delta m_F = 0$ if the r.f. field is parallel to H_0.

The only difference between the method described above and the ABMR method introduced by Rabi [13] is that the atoms are spin-polarized by laser interaction instead of magnetic deflection in the A magnet. Such a possibility of replacing deflecting inhomogeneous magnetic field by optical pumping had been pointed out several years ago [17-19]. Moreover, just as in the flop-in geometry in classical ABMR method, positive signals on a zero background could be achieved, provided that the hyperfine component to be matched with the laser frequency is suitably chosen.

3. **Apparatus.** — The apparatus is almost the same as the one used for previous measurements on sodium isotopes and a detailed description can be found in reference [4]. We report here only the specific modifications introduced for this experiment.

A carbon and iridium powder target is placed inside a high temperature oven. When bombarded by the 20 GeV proton beam from the proton synchrotron (P.S.) at C.E.R.N., the many produced nuclei, once thermalized, effuse from the oven and form an atomic beam (see Fig. 1*a*). The laser system consists in a commercial C.W. single mode dye laser (CR 599) pumped by a Kr^+ laser (CR 3 000 K). Its frequency is scanned step by step and servocontrolled by a « sigmameter » [20]. The proper dye to be used at the

wavelength of the potassium D_1 line (λ = 770 nm) is oxazine 750. One watt r.f. power, continuously tunable in the range 10 MHz-1 GHz is delivered by a commercial r.f. generator (Rohde and Schwartz SMS 0.4-1 040 MHz) followed by a linear wide band amplifier.

Special care has been taken in the design of the interaction region, schematically shown in figure 1*b*. Two Helmholtz coils produce a homogeneous static magnetic field H_c parallel to the laser beam while the r.f. magnetic field is produced by a single-turn copper loop centred on the atomic beam and perpendicular to it. Therefore the r.f. magnetic field seen by the atoms is parallel to the atomic beam direction. The whole interaction region is surrounded by a magnetic shield made of Armco iron and mu-metal sheet. In the direction of the sixpole magnet a hole is bored in this shield, with such a diameter that the magnetic stray field from the sixpole magnet is sufficient to avoid nonadiabatic transitions. This inhomogeneous stray field $\mathbf{H}_s$, is very weak at the r.f. coil (typically 0.1 G) and pointing approximately parallel to the atomic beam. The total static magnetic field is $\mathbf{H}_0 = \mathbf{H}_c + \mathbf{H}_s$; for $H_c \gg H_s$ it is approximately perpendicular to the atomic beam and homogeneous; for $H_c = 0$ it is quite parallel to the atomic beam and strongly inhomogeneous.

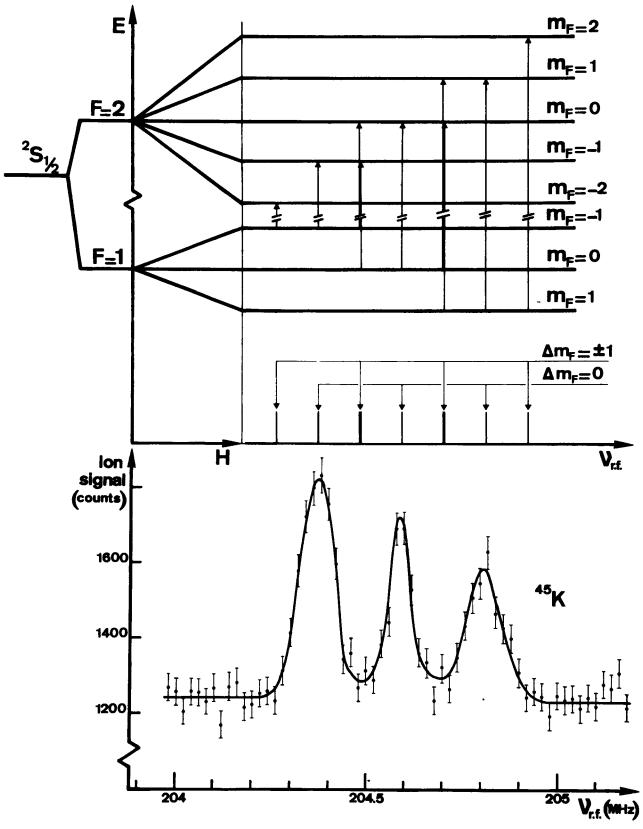

Fig. 2. — Zeeman splitting of the $\Delta F = 1$ hyperfine transition for ^{45}K. The recording shown in the lower part of the figure has been obtained with a static field parallel to the r.f. field (see text). Therefore resonances are observed only for the $\Delta m_F = 0$ components. Note that the central resonance is narrower than the two other ones which are broadened due to the strong inhomogeneity of the static magnetic field.

Figure 2 presents a recording of the Zeeman components of the $\Delta F = 1$ r.f. transition for ^{45}K. It has been obtained without d.c. current in the Helmholtz coils and therefore the static magnetic field H_0 is the stray field H_s. Since the r.f. field and H_0 are then parallel, only the $\Delta m_F = 0$ resonances occur. Moreover, the central resonance is narrower than the two other ones because it is field independent to first order while the two others are broadened due to the strong inhomogeneity of the static magnetic field.

4. **Experimental procedure.** — Several operations are successively carried out for measuring the hyperfine splitting of the ground state. First an approximate value of this hyperfine splitting is obtained by laser spectroscopy together with the sign of the nuclear magnetic moment as explained in paragraph 2. Then the static field $\mathbf{H}_c$ is set to a value in the range 0.5-1 gauss so that the amplitude of $\mathbf{H}_0$

$$(H_0 = \sqrt{H_c^2 + H_s^2})$$

is roughly known. $\mathbf{H}_0$ is nearly, but not exactly, perpendicular to the atomic beam, i.e. the direction of the r.f. magnetic field. Therefore the $\Delta m_F = \pm 1$ resonances are easily excited with low r.f. power while for the $\Delta m_F = 0$ resonances more r.f. power must be applied to the loop. Figure 3 presents all the Zeeman components of the $\Delta F = 1$ transition in the case of ^{43}K ($I = 3/2$, $\mu_I > 0$). All the components, except $(1, -1) \leftrightarrow (2, -2)$, correspond to a change in m_J and are then observable. Moreover, these resonances are approximately equidistant and the frequency spacing, essentially related to H_0, is roughly known. Such a situation is time-saving in the search of a first resonance because it permits to reduce the frequency range to be scanned. When one component has been found it becomes easy to record the other ones. All the components of the Zeeman splitting of ^{43}K have been recorded although this procedure was not necessary because the hyperfine structure of the ground state was already known with an accuracy of 50 kHz [21]. The result is shown on figure 3. The same procedure was repeated for ^{44}K.

It is well known in ABMR technique that one of the components is only second order magnetic field dependent, i.e. $(I + 1/2, 0) \leftrightarrow (I - 1/2, 0)$ for the odd isotopes and the unresolved doublet

$$\{(I+1/2, 1/2) \leftrightarrow (I-1/2, -1/2) \quad (I+1/2, -1/2) \leftrightarrow (I-1/2, 1/2)\}$$

for the even isotopes [22]. The last step of the experimental procedure is to identify clearly this component and to precisely measure its frequency. Since this component does not practically depend on the magnetic field H_0 it can be identified by changing the magnetic field amplitude.

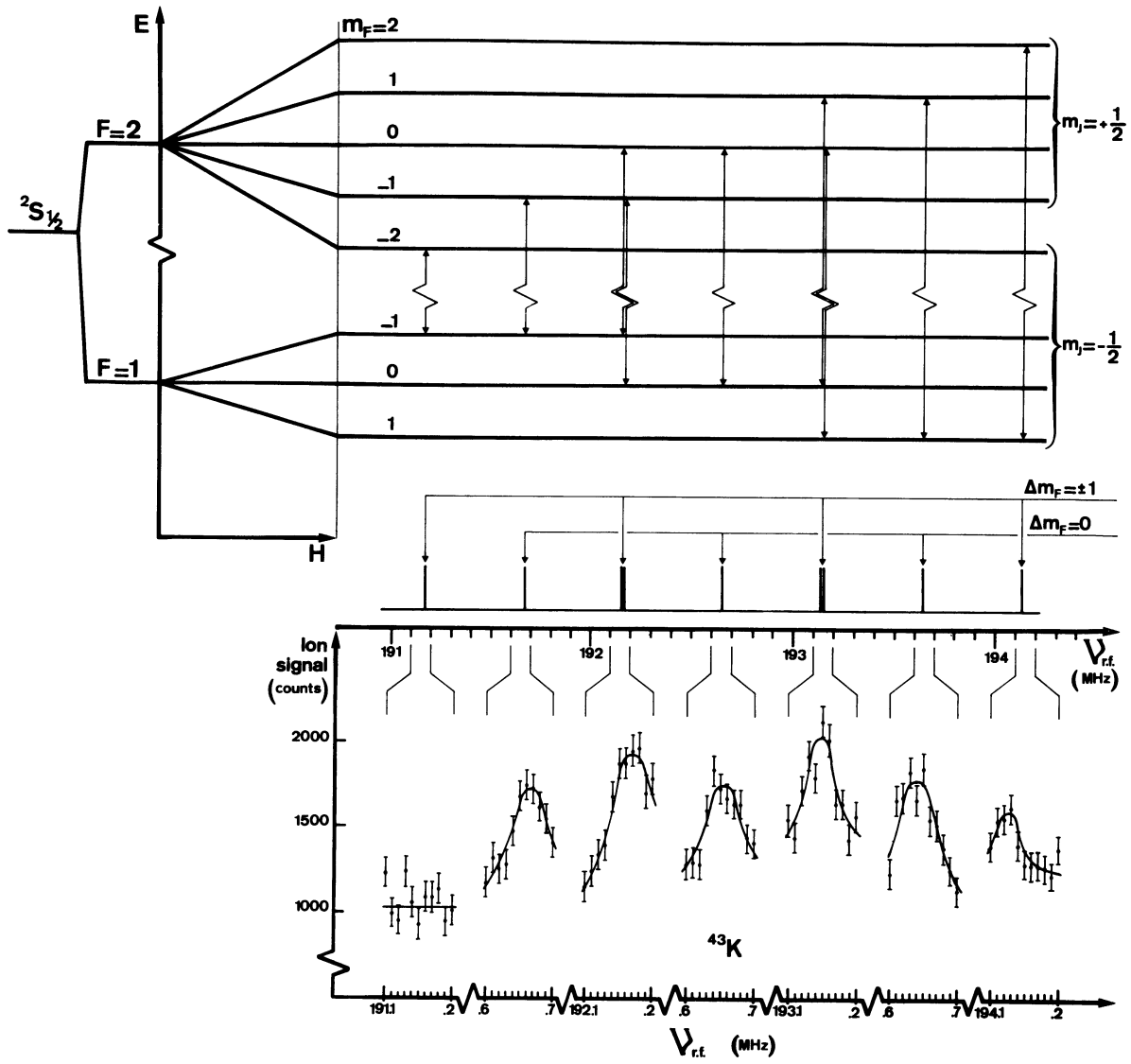

Fig. 3. — Zeeman splitting of the $\Delta F = 1$ hyperfine transition in the case of ^{43}K ($I = 3/2$, $\mu_I > 0$). All the Zeeman components, except $(1, -1) \leftrightarrow (2, -2)$, correspond to a change in m_J and are therefore observable. In the lower part of the figure the frequency scale is strongly expanded for each recorded resonance.

5. **Results and discussion.** — Figure 4 presents a precise recording of the first order field independent doublet of ^{44}K. To get the exact hyperfine structure in zero field one has to take into account the second order correction. An accurate value of the magnetic field amplitude H_0 can be deduced by a least-square fit from the experimental frequency spacings between the Zeeman components of the r.f. transition. This fit gives $H_0 = 0.704$ G (respectively 0.700 G) for ^{43}K (respectively ^{44}K) with an accuracy better than 1 %. Taking into account this value of the magnetic field H_0 the second order correction is calculated for both isotopes (− 10 kHz for ^{43}K and − 2 kHz for ^{44}K). The deduced values of the ground state hyperfine structures are indicated in table I.

For ^{43}K, our measurement is in agreement with a less precise value quoted in the literature [21]. Our 3 kHz error bar for ^{43}K and ^{44}K is estimated from the consistency between several measurements of the same resonance. We have also measured the well known hyperfine structures of ^{45}K [23] and ^{20} Na [24]. The 6 kHz error bar reflects the shorter time devoted to these measurements. Our results, reported in table I, are in very good agreement with earlier, more precise, results.

As pointed out, the present method is comparable to the classical ABMR method working in the flop-in arrangement. Our apparatus presents the advantage of simplicity and its length is greatly reduced compared to a classical ABMR apparatus : the distance from the oven to the entrance of the sixpole magnet, which corresponds to the length of the A and C regions of an ABMR apparatus, is only seven centimeters (see Fig. 1*a*). This short distance and the focussing properties of the sixpole magnet provide a high acceptance suitable for the study of very rare isotopes. Never-

Table I. — *Measured ground state hyperfine structures.*

Isotope	$\Delta\nu_{H.F.S.}$ ($F = I - 1/2 \leftrightarrow F = I + 1/2$) [MHz]		$A(^2S_{1/2})$ [MHz]
	our results	other results	deduced from the most precise result
^{43}K	192.648 4 (30)	192.64 (5) ([a])	96.324 2 (15)
^{44}K	− 946.718 (3)		− 378.687 2 (12)
^{45}K	204.590 (6)	204.587 3 (15) ([b])	102.293 6 (8) ([b])
^{20}Na	276.858 (6)	276.855 (3) ([c])	110.742 0 (12) ([c])

([a]) Ref. [21].
([b]) Ref. [23].
([c]) Ref. [24].

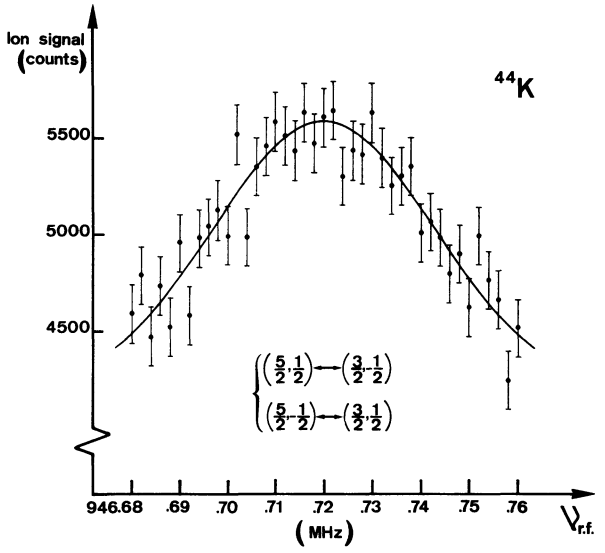

Fig. 4. — Typical recording of the resonance curve corresponding to the first order field independent unresolved doublet for ^{44}K.

theless it is difficult to deduce from this experiment our limit of sensitivity. Production rates of ^{44}K and ^{45}K were about 2.5 × 10^7 atoms/proton pulse (proton pulse intensity : 10^{13} protons) but they do not represent the lower limit (see the recording of ^{45}K on figure 2). Moreover the on-line produced potassium isotopes were unfavourable cases for two reasons :

— For ^{43}K and ^{44}K, signals were perturbed by an important background due to natural calcium isobars coming from the heated ion source. This background has been subtracted on the recordings but it contributes to the statistical noise.

— For ^{44}K, the nuclear magnetic moment is negative; this is unfavourable since it is then impossible to optically pump all the atoms into defocussed states and therefore, the flop-in resonances do not occur on a zero background.

One can get an idea of the sensitivity of our method from the limit of sensitivity obtained with the same apparatus in optical laser spectroscopy work. Optical spectroscopy has been performed with production rates as low as 10^4 atoms/proton pulse for sodium at the proton synchrotron of C.E.R.N. [25] and 10^5 atoms/s for rubidium at the ISOLDE mass separator (C.E.R.N.) [5]. In our method, the r.f. resonance signals are of course weaker than the optical resonance signals because the Zeeman degeneracy is removed but the loss in sensitivity due to this fact is, in all cases, certainly less than a factor of ten. Therefore our method can be used with isotope production rates higher than 10^6 atoms/s. This limit corresponds to the one obtained with a very efficient ABMR apparatus [26] especially designed to work with atomic beams of rare isotopes.

References

[1] BONN, J., KLEMPT, W., NEUGART, R., OTTEN, E. W. and SCHINZLER, B., *Z. Phys.* **A 289** (1979) 227.

[2] BEKK, K., ANDL, A., GÖRING, S., HANSER, A., NOWICKI, G., REBEL, H. and SCHATZ, G., *Z. Phys.* **A 291** (1979) 219.

[3] BERGMANN, E., BOPP, P., DORSCH, C., KOWALSKI, J., TRÄGER, F. and ZU PUTLITZ, G., *Z. Phys.* **A 294** (1980) 319.

[4] HUBER, G., TOUCHARD, F., BÜTTGENBACH, S., THIBAULT, C., KLAPISCH, R., DUONG, H. T., LIBERMAN, S., PINARD, J., VIALLE, J. L., JUNCAR, P. and JACQUINOT, P., *Phys. Rev.* **C 18** (1978) 2342.

[5] THIBAULT, C., TOUCHARD, F., BÜTTGENBACH, S., KLAPISCH, R., DE SAINT SIMON, M., DUONG, H. T., JACQUINOT, P., JUNCAR, P., LIBERMAN, S., PILLET, P., PINARD, J., VIALLE, J. L., PESNELLE, A. and HUBER, G., *Phys. Rev.* **C 23** (1981) 2720.

[6] THIBAULT, C., TOUCHARD, F., BÜTTGENBACH, S., KLAPISCH, R., DE SAINT SIMON, M., DUONG, H. T., JACQUINOT, P., JUNCAR, P., LIBERMAN, S., PILLET, P., PINARD, J., VIALLE, J. L., PESNELLE, A. and HUBER, G., *Nucl. Phys.* **A 367** (1981) 1.

[7] LIBERMAN, S., PINARD, J., DUONG, H. T., JUNCAR, P., PILLET, P., VIALLE, J. L., JACQUINOT, P., TOUCHARD,

F., BÜTTGENBACH, S., THIBAULT, C., DE SAINT SIMON, M., KLAPISCH, R., PESNELLE, A. and HUBER, G., *Phys. Rev.* **A 22** (1980) 2732.

[8] TOUCHARD, F., BÜTTGENBACH, S., GUIMBAL, P., KLAPISCH, R., DE SAINT SIMON, M., SERRE, J. M., THIBAULT, C., DUONG, H. T., JUNCAR, P., LIBERMAN, S., PINARD, J., VIALLE, J. L. and HUBER, G., to be published in *Phys. Lett. B* (1982).

[9] BENDALI, N., DUONG, H. T. and VIALLE, J. L., *J. Phys. B* **14** (1981) 4231.

[10] BUCK, P. and RABI, I. I., *Phys. Rev.* **107** (1957) 1291.

[11] NEY, J., *Z. Phys.* **223** (1969) 126.
NEY, J., REPNOW, R., BUCKA, H. AND SVANBERG, S., *Z. Phys.* **213** (1968) 192.

[12] SCHMIEDER, R. W., LURIO, A. and HAPPER, W., *Phys. Rev.* **173** (1968) 76.

[13] RABI, I. I., ZACHARIAS, J. R., MILLMAN, S. and KUSCH, P., *Phys. Rev.* **53** (1938) 318.

[14] FULLER, G. H., *J. Phys. Chem. Ref. Data* **5** (1976) 835.

[15] PENSELIN, S., MORAN, T., COHEN, V. W. and WINKLER, G., *Phys. Rev.* **127** (1962) 524.

[16] DUONG, H. T. and VIALLE, J. L., *Opt. Commun.* **12** (1974) 71.

[17] KASTLER, A., *J. Physique Radium* **11** (1950) 255.
DEMBCZYNSKI, J., ERTMER, W., JOHANN, U., PENSELIN, S. and STINNER, P., *Z. Phys.* **A 291** (1979) 207.

[18] BUCKA, H., *Z. Phys.* **191** (1966) 199.

[19] ARDITI, M. and PICQUÉ, J. L., *J. Physique Lett.* **41** (1980) L-379 and *C.R. Hebd. Séan. Acad. Sci.* Paris **B 290** (1980) 461.

[20] JUNCAR, P. and PINARD, J., *Opt. Commun.* **14** (1975) 438.

[21] PETERSEN, F. R., EHLERS, V. J., EWBANK, W. B., MARINO, L. L. and SHUGART, H. A., *Phys. Rev.* **116** (1959) 734.

[22] RAMSEY, N. F., *Molecular Beams* (Clarendon Press, Oxford) 1956, p. 84.

[23] GARRETT, G. J., JACKSON JR., A. D. and AMES, O., *Phys. Rev.* **161** (1967) 1152.

[24] SCHWEICKERT, H., DIETRICH, J., NEUGART, R. and OTTEN, E. W., *Nucl. Phys.* **A 246** (1975) 187.

[25] KLAPISCH, R., paper presented at the *7th Inter. Conf. on Atomic Physics*, (Cambridge, Mass. U.S.A.) August 4-8 (1980).

[26] EKSTRÖM, C. and LINDGREN, I., in *Atomic Physics*, Vol. **5**, eds. R. Marrus, M. Prior and H. Shugart (Plenum Press, New York) 1977, p. 201.